REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comment regarding this burden estimates or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for information Operations and Reports, 1215 Jefferson Davis Highway, Surte 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.			
		AND DATES COVERED	
4. TITLE AND SUBTITLE	7/22/00	Final 9/1/9	6 to 2/28/00 5. FUNDING NUMBERS
Combined Far-Infrared & Optical Probes of Semiconductor Quantum Structures			
6. AUTHOR(S)			DAAH04-96-1-0414
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7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)			8. PERFORMING ORGANIZATION REPORT NUMBER
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3003 S. State St.			
Ann Arbor, MI 48109-1287 9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)			10. SPONSORING / MONITORING
			AGENCY REPORT NUMBER
U.S. Army Research Office P.O. Box 12211			
Research Triangle Park, NC 27709-2211			ARO 35827.10-PH
AL OURDING MENTARY NOTES			
11. SUPPLEMENTARY NOTES  The views opinions and/or findings contained in this report are those of the author(s) and should not be construed as			
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.			
12a. DISTRIBUTION / AVAILABILITY STATEMENT			12 b. DISTRIBUTION CODE
Approved for public release; distribution unlimited.			
13. ABSTRACT (Maximum 200 words)			
We report on a program in which we have developed a terahertz time-resolved spectroscopy system with electro-			
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depth study to date of carrier capture and relaxation dynamics in self-organized III-V quantum dots. In a major			
breakthrough, we have made the first direct, spectroscopic observation of the phonon bottleneck, and also the first			
observation of the fast electron-hole scattering that occurs when both species of carrier are captured in the same			
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14. SUBJECT TERMS 2000112 107			15. NUMBER IF PAGES
14. SUBJECT TERMS 20001122 083			
			16. PRICE CODE
	. SECURITY CLASSIFICATION	19. SECURITY CLASSIFIC	ATION 20. LIMITATION OF ABSTRACT
OR REPORT UNCLASSIFIED	OF THIS PAGE UNCLASSIFIED	OF ABSTRACT UNCLASSIFIED	, , , , , , ,

# Combined Far-Infrared and Optical Probes of Semiconductor Quantum Structures

## **Final Report**

submitted to Dr. Henry Everitt
Physics Division
US Army Research Office

July 22, 2000

Principal Investigator:

Prof. Theodore B. Norris Center for Ultrafast Optical Science EECS Department, University of Michigan 2200 Bonisteel Blvd, Ann Arbor, MI 48109-2099 (313) 764-9269

#### **Foreward**

Our goal in this three-year program was to develop combined femtosecond optical and far-infrared probes and apply them to the study of carrier dynamics in semiconductor nanostructures relevant to optoelectronic device operation. With the support of this program, we have developed a terahertz time-resolved spectroscopy system with electrooptic generation and detection, incorporating a femtosecond optical pump pulse suitable for studying the dynamics of impulsively photo-generated carriers using the THz probe. In the course of this work, we have developed an entirely new way of generating tunable coherent narrow-band (multi-cycle) THz waveforms using femtosecond pulses. The technique exploits the group velocity mismatch between optical and THz waves in periodically-poled lithium niobate (PPLN), and generates tunable far-infrared pulses suitable for semiconductor spectroscopy. We have also utilized all-optical femtosecond pump-probe spectroscopy to perform the most in-depth study to date of carrier capture and relaxation dynamics in self-organized III-V quantum dots. In a major breakthrough, we have made the first direct, spectroscopic observation of the phonon bottleneck, and also the first observation of the fast electron-hole scattering that occurs when both species of carrier are captured in the same quantum dot; this electron-hole scattering circumvents the phonon bottleneck and enables fast relaxation in bipolar device structures. Additionally, we have directly observed for the first time the tunnel coupling of vertically aligned quantum dots, and performed the first study of the temperature dependence of carrier capture in these systems.

## I. Statement of the Problems Studied

Our principal accomplishments over the course of the three-year program were in two general areas: (A) Development of a novel optoelectronic THz probe system, and (B) time-resolved carrier dynamics in self-organized quantum dots. In the next section I briefly outline the main achievements and their significance. I have not included significant technical detail here; instead, I have attached a series of reprints and preprints of our publications which describe the technical approach and data in detail, and the interested reader is referred to those papers.

## II. Summary of the Most Significant Results

## A. Optoelectronic THz Probe Development

As discussed in our original proposal, there are a number of new capabilities in time-resolved semiconductor studies which would be made possible by a combined optical/THz pump-probe system. The experimental technique consists of a subpicosecond optical pump pulse followed by a far-infrared THz pulse at variable delay. The THz pulse is generated optoelectronically from the same laser as the optical pump pulse. The optical pump generates population in the sample, while the THz pulse probes the transient induced excited-state absorption, which is proportional to the photoexcited population. This technique can be applied to a variety of issues in excited-state dynamics

in semiconductor quantum structures. One general set of problems concerns the measurements of dark states, such as k>0 excitons and cavity polaritons, or dipoleforbidden exciton spin states. Because the scattering rates of excitons are comparable to radiative recombination rates, many fundamental questions regarding the radiative dynamics of excitons in quantum wells and microcavities have not been definitively answered. The combined optical-THz probe will allow us to determine directly for the first time the dynamics of the total population of excitons in these structures, not just the k=0 dipole-allowed population accessed by all-optical experiments. Additionally, such experiments will allow us to test recently developed many-body theories which call into question the whole concept of excitonic luminescence which has served as the basis for our understanding of light emission from semiconductors (in particular, the formation of excitons from free electron-hole pairs needs to be revisited). The second general set of problems to be addressed concerns the relaxation of electrons and holes within quantum well subbands or between quantum dot excited states. The combined optical-THz probe can be used to directly study the relaxation rates in these materials, and to determine THz gain and loss dynamics in far infrared laser and detector structures.

With support from our ARO program, we have developed a pump-probe station to perform the optical/THz experiments. In the first year of the program, I hired a new graduate student research assistant, Tim Meade, who was responsible for building the system and performing initial experiments. Tim was a new student, so most of his effort was in learning how THz measurements are made and building the experimental setup. He initially succeeded in obtaining real-time THz signals using LT-GaAs-based photoconductive generation and detection. As the bandwidth of the photoconductive THz system was limited to about 1 THz (limited partly by our ability to obtain good high-resistivity LT-GaAs with sufficiently short carrier lifetimes), and our probes of exciton and free-carrier dynamics requires a bandwidth of greater than 2 THz, he then implemented electro-optic (ZnTe-based) THz emission and detection. The ZnTe-based system gives bandwidths easily in excess of 2 THz, but is not ideal for probing the exciton dynamics (for which the 1s-2p exciton transition has a resonance near 2 THz) The reason is that the ZnTe signal, while broadband, is also dispersed, reducing the time resolution without concentrating spectral density near the 1s-2p transition.

In order to concentrate the spectral density of the THz probe, it is necessary to generate multi-cycle THz waveforms, thus yielding a narrower bandwidth. One of our most significant breakthroughs in this program was the invention of a new way of generating shaped, multi-cycle, narrowband THz waveforms using femtosecond lasers.

The basic idea is illustrated in figure 1. A short (100 fs) optical pulse is propagated through periodically-poled Lithium Niobate (PPLN). In each domain, the sign of  $\chi^{(2)}$  is reversed, so that the optical pulse generates a THz nonlinear polarization via optical rectification which oscillates in sign as the pulse propagates. The THz wave propagates slower than the optical pulse, so the technique effectively utilizes the group-velocity walkoff between the optical and THz pulses. The THz waveform is essentially given by the domain structure of the PPLN crystal. Detailed descriptions of the technique and the main results may be found in the (p)reprints attached to this report. So far, this work has been published in three conference proceedings, two Applied Physics Letters, with an additional Appl. Phys. Lett. submitted.

An example of the data is shown in figure 2, for a 1.2-mm PPLN crystal at low temperature, with a domain length of 30  $\mu$ m, corresponding to a peak frequency of 1.8 THz.

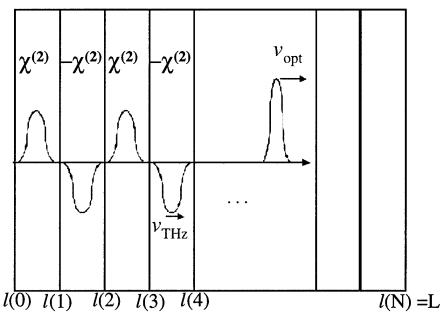


Figure 1. Schematic of THz waveform generation in PPLN. The optical pulse generates an oscillating nonlinear polarization via optical rectification as it propagates through the crystal.

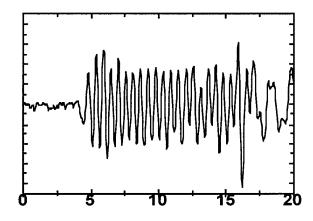


Figure 2. THz waveform emitted from a 1.2-mm long PPLN crystal with 30-micron domain length. The peak frequency is at 1.8 THz with a bandwidth of 70 GHz.

The significance of this new technique is that it allows the generation of narrow-band THz pulses using femtosecond pulses, and not just the well-established single-cycle pulses. For example, using a 7.2-mm PPLN crystal at low temperature, we have

generated waveforms at 1.8 THz with a bandwidth of only 18 GHz. The scheme easily enables simple tunability across the few-THz range. To our knowledge, these THz pulses have the highest spectral brightness of any THz generation technique yet demonstrated.

Now that we are able to generate bright THz pulses tunable to any resonance in the few-THz regime, we are finally in a position to carry out the combined optical-THz experiments on semiconductor nanostructures. The development stage of the project lasted significantly longer than we expected, but the work resulted in a significant breakthrough in the generation of coherent THz electromagnetic radiation.

#### B. Carrier Dynamics in Self-Organized Quantum Dots

Our second major effort in this program has been in the study of self-organized InGaAs/GaAs quantum dots. This quantum dot system is one of the most promising for applications (such as mid- and far-IR detectors and emitters) due to their high quality relative to other dot systems. Such devices would operate on optical transitions between the quantum dot levels ("intersubband" transitions), and one of the principal physical effects determining device operation will be the carrier relaxation rates between the levels.

One of the outstanding questions regarding quantum dots has been the possibility of a "phonon bottleneck" in the relaxation, which would result in very long inter-level relaxation times for dots with sufficiently large interlevel energy spacings. The effect of this slow relaxation would be detrimental for the operation of high-speed interband lasers at optical frequencies. On the other hand, the presence of a phonon bottleneck would be advantageous for intersubband, FIR devices. For FIR lasers, a slow interlevel relaxation rate would facilitate the generation of a population inversion. For FIR detectors, the responsivity is directly proportional to the ratio of the upper state or continuum lifetime to the collection time (transit time across the device). Clearly an improvement in the upper state lifetime is desirable, and this can be accomplished by utilizing QD's instead of OW's.

In the past 3 years, we have carried out an extensive series of experiments using interband pump-probe time-resolved spectroscopy to determine the dynamics of carriers photoinjected into InGaAs quantum dots. I give here a brief overview of our main results and their significance; further technical details can be found in the attached (p)reprints. These experiments provide the first direct measurement of electron and hole relaxation in the self-organized InGaAs QD system, and required the refinement of femtosecond white-light spectroscopy to enable the measurement of differential transmission (DT) spectra in the 1-micron spectral region with the unprecedented sensitivity of a few parts in 10<sup>6</sup>.

Our first published results concerned the relaxation of electron-hole pairs within a quantum dot, and the capture of pairs into the dot states [Sosnowski Phys. Rev. B 1998]. In this first series of experiments, a femtosecond laser pulse was used to directly inject electrons and holes into their respective n=2 levels. A time-delayed tunable femtosecond probe pulse was then used to determine the carrier populations in both the ground and excited (n=2) states. The experiments were performed at low carrier density (less than one electron-hole pair per dot); in this case the DT spectrum could be simply interpreted in terms of the electron and hole distribution functions  $f_e$  and  $f_h$ :  $DT = \alpha(1 - f_e - f_h)$ ,

where  $\alpha(\omega)$  is the QD absorption spectrum. We observed a two-component decay of the n=2 population with a corresponding two-component rise of the n=1 population. Following theoretical modeling from Prof. Jasprit Singh's group here at Michigan, we ascribed the faster component (600 fs) to the hole relaxation and the slower component (5 ps) to the electron relaxation. The electronic relaxation rate we observe is orders of magnitude faster than one would expect based on phonon scattering; thus no phonon bottleneck limits the relaxation in this system. Instead, an electron can scatter

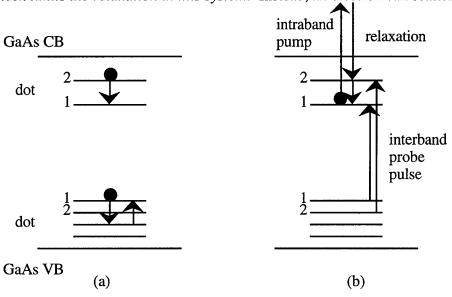


Figure 3 (a) Electron-hole scattering process; an electron scatters from n=2 to 1 by exciting a hole from 1 to 2. The hole subsequently loses its energy to the lattice. (b) Scheme for future experiments probing relaxation processes in unipolar quantum dot devices; an intraband (mid- to far-IR) pump pulse excites electrons from n-doped quantum dots, and the relaxation is subsequently probed via an interband optical pulse.

with a hole in the same quantum dot in an Auger type process. The rates we observed are quite close to those predicted by a self-consistent model of electron-hole scattering in quantum dots, assuming the holes remain in thermal equilibrium with the lattice (the holes are more strongly coupled to the phonons). In this process, an electron relaxes from the n=2 to the n=1 level in the QD by kicking a hole from its ground state to an excited state, as shown schematically in figure 3(a). In an isolated QD, such a process could not occur, since the electron and hole energy level spacings do not match, and the δ-function density of states would forbid this scattering process as it would require violation of energy conservation. When coupling of the holes to the lattice phonons is included, the resulting broadening of the hole levels relaxes the energy conservation requirement, thus opening up the electron-hole scattering process. It is important to note that this electron-hole scattering process dominates even though the pair density is quite low (less than one pair per dot), since the scattering occurs within the quantum dot.

In the second series of experiments, the pump pulse was tuned to inject electronhole pairs into the GaAs barrier region surrounding the dots, and the population of the n=1 and 2 levels was monitored with the probe. We directly time-resolved the carrier capture into the dots (approximately 2.8 ps into the excited state, and a net relaxation time into the ground state of about 10 ps). These experiments were the first direct femtosecond time-resolved measurements of carrier capture and relaxation in the InGaAs self-organized dot system (the measurements are very difficult due to the low absorption in the dots as compared to, say, quantum wells), and the results are particularly relevant to the operation of interband diode lasers (or indeed any other bipolar device). Although the measurements were all performed at low density, we have found that our pump-probe

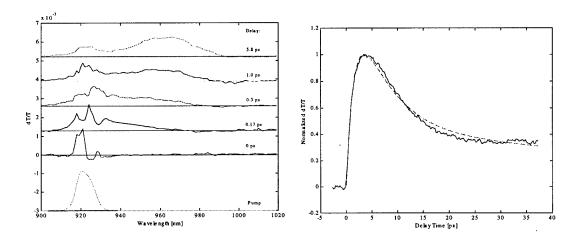


Figure 4. On the left (a) is an example of time-resolved DT spectra showing the rapid energy redistribution of carriers in the quantum dot excited state due to interdot coupling. On the right (b) is an example of the temporal dynamics of the n=2 state when the barrier region is pumped; the long tail may be due to long lived (bottlenecked) electrons nongeminately captured into dots without holes.

results are reasonably consistent with the measurements of laser modulation bandwidths in the same system [Bhattacharya 1999], and thus the phonon bottleneck does not prevent fairly high modulation rates in diode laser operation.

The story is expected to be quite different for unipolar devices, such as mid-IR emitters and detectors. In this case, since only electrons are present in the system, the electron-hole scattering mechanism can not operate, and only phonons can mediate the relaxation if the electrons are all in quantum dot excited states. Some of our most recent measurements [Urayama Appl. Phys. Lett., Phys. Rev. Lett. 2000] indicate (1) that the excited states of the self-organized dots are coupled, and (2) that there may be a relaxation bottleneck in dots containing only electrons.

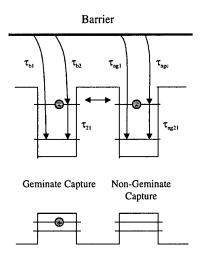
In one series of experiments, we have tuned the excitation to inject electron-hole pairs into the n=2 level of the InGaAs dots, and using a white light probe, measured the entire differential transmission *spectrum* as a function of time delay. An example of the data is shown in figure 2(a). The pump is tuned to 920 nm, which excites the subset of the QD's within the inhomogeneous dot size distribution that have n=2 electron-hole transitions resonant with the pump. The white light probe allows determination of the entire carrier distribution functions via  $DT = \alpha(1 - f_e - f_h)$ .

In the absence of coupling between QD's, the pump spectrum should "burn a hole" in the probe absorption spectrum, so at early probe delay times the DT spectrum should be the same as the pump spectrum. As time evolves, and carriers relax from the n=2 to

the n=1 state, a "replica" spectral hole should appear in the DT spectrum around the n=1 transition. This is expected since the initially excited energy distribution is much narrower than the QD inhomogeneous spectrum and the relaxation of a given e-h pair occurs within a single QD.

As is clear from our preliminary data shown in figure 4(a), this is not what is observed experimentally. The spectral hole burned by the pump lasts only about 100 fs

# Temperature Dependent Carrier Dynamics In In<sub>0.4</sub>Ga<sub>0.6</sub> As Quantum Dots



Rate Equation Model

Figure 5. Schematic diagram of the energy levels and relaxation processes in the self-organized quantum dot structure, showing the various processes included in the rate equation model for the DT dynamics.

(the oscillatory structure in the DT spectrum around the pump energy is due to probe pulse interference with scattered pump light). On the time scale of a few hundred fs, the DT spectrum shows a rapid broadening (particularly to lower energies). This can only occur if the QD n=2 excited states are coupled. We have performed preliminary investigations of the carrier density dependence of the rapid initial broadening and find it to be independent of density over the experimentally accessible range, indicating that carrier-carrier scattering is unlikely to be the physical cause of this broadening. Instead, we propose that the dot levels are coherently coupled, and thus the rapid evolution of the DT spectrum is due to the motion of the spatial wavepacket excited by the pump pulse. The coherent coupling arises in our sample because there are 4 layers of quantum dots in the active region, and it is well known that the quantum dots in the different layers are vertically aligned (we have confirmed this using cross-sectional TEM measurements in collaboration with Prof. Rachel Goldman here at Michigan). Thus the coherent coupling arises because of overlap of the electronic wavefunctions of vertically aligned quantum dots. Note that decay rather than oscillatory dynamics are observed because of the broad inhomogeneous distribution of QD sizes. The strong vertical coupling between dots has

important implications for devices which are designed to exploit a  $\delta$ -function-like density of states as expected in QD's.

Whereas the vertical coupling of aligned self-organized quantum dots is strong (leading to tunneling times between dots of only a few hundred fs), the horizontal coupling is calculated to be orders of magnitude weaker (due to the much smaller wavefunction overlap). Thus an interesting possibility opens up in electronic capture experiments: if an electron-hole pair is photoinjected into the continuum states in the barrier region above the quantum dots, then (a) the electron and hole may be captured into the same quantum dot (geminate capture), or (b) the electron and hole may be captured into laterally separate quantum dots (nongeminate capture). Geminately captured electrons can undergo electron-hole scattering leading to fast relaxation to the ground state (bypassing the phonon bottleneck). On the other hand, nongeminately captured electrons have no hole with which to scatter, and thus they remain in the quantum dot excited state for a long time due to the phonon bottleneck. Such a bottlenecked population would appear as a long tail in the decay of a differential transmission experiment.

The observation of this effect requires that differential transmission experiments be done at very low density, so that a significant fraction of electrons will be trapped into dots without holes. By making further improvements to our pump-probe system, we were able to obtain high signal-to-noise ratio while photoinjecting only one electron-hole pair per ten quantum dots, and we were thus able to see clearly the effect of nongeminate capture. In figure 4(b), we show some DT data showing the dynamics of the n=2 state when the barrier region is pumped. The rise is due to carrier capture, and the initial decay is due to relaxation from n=2 to 1 via electron-hole scattering as discussed above. The signal does not decay to zero, however, but shows a long tail. The data can be fit by a rate equation model which takes into account both geminate and nongeminate capture. A schematic of the processes included in the model is shown in figure 5; the hole is assumed to be captured into the left QD, and the electron may be captured into the same or a different dot. Coupling between the dots can be included as a parameter. The fit to the data is excellent. (It should be noted that the data can also be fit if a long-lived deep defect level contributes to the DT spectrum; additional experiments have ruled out this possibility.) If the capture model is correct, it shows that nongeminately captured electrons may stay in the excited state for of order 200 ps, i.e. nearly two orders of magnitude longer than geminately captured electrons. This time constant is probably not the phonon-assisted relaxation time, but rather is due to either inter-dot coupling or to thermal re-emission into the barrier followed by possible recapture into a dot containing a hole. Thus the excited state dynamics are thus temperature dependent; we are presently still analyzing the data and writing the results up for publication.

In the course of our 3-year program, we have thus achieved a fairly comprehensive understanding of the dynamics of electron-hole pairs in self-organized InGaAs quantum dots. We have shown that when electrons occupy the same quantum dot as holes, then the interlevel relaxation is fast, and the phonon bottleneck is bypassed. When the electrons are in separate quantum dots, then the Coulomb scattering is suppressed, and the relaxation is bottlenecked. When an electron is injected into a single quantum dot in a multi-layer structure, then the electron can tunnel quickly to other vertically aligned dots due to coherent electronic coupling.

These experiments are critical to the understanding of bipolar optoelectronic devices based on quantum dots. Clearly, further work is required to understand the dynamics in unipolar devices, where no holes are present. These experiments can be done using the scheme shown in figure 3(b), in which electrons in a doped quantum dot are excited by an infrared pulse; this will be a major focus of our work in the coming period.

#### III. List of Publications

- J. Urayama, T.B. Norris, J. Singh, and P. Bhattacharya, "Temperature Dependence of Carrier Capture and Relaxation in Self-Organized Quantum Dots," manuscript to be submitted to Appl. Phys. Lett.
- J. Urayama, T.B. Norris, J. Singh, and P. Bhattacharya, "Observation of Phonon Bottleneck in Qauntum Dot Electronic Relaxation," submitted to Phys. Rev. Lett.
- Y.-S. Lee, T. Meade, M. DeCamp, T.B. Norris, and A. Galvanauskas, "Temperature Dependence of Narrow-Band Terahertz Generation from Periodically-Poled Lithium Niobate," to be published in <u>Appl. Phys. Lett</u>.
- Y.-S. Lee, T. Meade, V. Perlin, H. Winful, T.B. Norris, and A. Galvanauskas, "Generation of Narrow-Band Terahertz Radiation via Optical Rectification in Periodically-Poled Lithium Niobate," Appl. Phys. Lett. 76, 2505 (2000).
- J. Urayama, T.B. Norris, B. Kochman, J. Singh, and P. Bhattacharya, "Evidence of Interdot Electronic Tunneling in Vertically Coupled In0.4Ga0.6As Self-Organized Quantum Dots," <u>Appl. Phys. Lett.</u> 76, 2394 (2000).
- P. Bhattacharya, K. Kamath, J. Singh, D. Klotzin, J. Phillips, H.-T. Jiang, N. Chervela, T. Norris, and T. Sosnowski, "In(Ga)As/GaAs Self-Organized Quantum Dot Lasers: DC and Small-Signal Modulation Properties," IEEE Trans. Electron Devices 46, 871 (1999). ). [This paper received the Paul Rappaport Award for best paper in an IEEE Electron Devices Society publication, 1999.]
- T. Sosnowski, J. Urayama, T.B. Norris, H. Jiang, J. Singh, K. Kamath, J. Phillips, and P. Bhattacharya, "Ultrafast Carrier Capture and Relaxation in InGaAs and InAs Self-Organized Quantum Dots," Trends in Optics and Photonics Vol. 18, Radiative Processes and Dephasing in Semiconductors, D.S. Citrin, ed., (Optical Society of America, 1998), p.30.
- T.S. Sosnowski, T.B. Norris, H. Jiang, J. Singh, K. Kamath, and P. Bhattacharya, "Rapid Carrier Relaxation in In0.4Ga0.6As/GaAs Quantum Dots Characterized by Differential Transmission Spectroscopy," Phys. Rev. B Rapid Communications 57, R9423 (1998).
- T.S. Sosnowski, T.B. Norris, H. Jiang, J. Singh, K. Kamath, and P.Bhattacharya, "Ultrafast Carrier Capture and Relaxation in InGaAsSelf-Organized Quantum Dots," <u>Optics and Photonics News 9</u>, 47 (1998).
- T. Meade, Y.-S. Lee, V. Perlin, H. Winful, T.B. Norris, and A. Galvanauskas, "Generation of Narrow-Band Terahertz Pulses by Optical Rectification in Periodiaclly-Poled Lithium Niobate," presented at the Ultrafast Phenomena Conference, Charleston, 2000.
- T.B. Norris, Y.-S. Lee, T. Meade, M. DeCamp, C. Herne, V. Perlin, H. Winful, and A. Galvanauskas, "Narrow-Band THz Waveforms from Optical Rectification of Femtosecond Optical Pulses in Periodically Poled Lithium Niobate," postdeadline paper presented at the Conference on Lasers and ElectroOptics, San Francisco, May 2000.
- J. Urayama, T.B. Norris, K. Kamath, and P. Bhattacharya, "Evidence of Interdot Carrier Coupling in In0.4Ga0.6As Self-Assembled Quantum Dots," paper QMJ5, Quantum Electronics and Laser Science Conference, San Francisco, May 2000.
- T. Meade, Y.-S. Lee, V. Perlin, H. Winful, T.B. Norris, and A. Galvanauskas, "Generation of Narrow-Band Terahertz Pulses by Optical Rectification in Periodically-Poled Lithium Niobate," presented at the 11<sup>th</sup> Int. Symp. on Space Terahertz Technology, Ann Arbor, 2000.

- T.S. Sosnowski, J. Uruyama, T.B. Norris, H. Jiang, J. Singh, K. Kamath, and P.Bhattacharya, "Ultrafast Carrier Capture and Relaxation in InGaAsSelf-Organized Quantum Dots," paper QThC4, International Quantum Electronics Conference, San Francisco, 1998.
- T.S. Sosnowski, T.B. Norris, H. Jiang, J. Singh, K. Kamath, and P.Bhattacharya, "Ultrafast Carrier Capture and Relaxation in InGaAsSelf-Organized Quantum Dots," paper RMB2, OSA Workshop on Radiative Processes and Dephasing in Semiconductors, Coeur d'Alene, 1998.
- K. Kamath, H. Jiang, D. Klotzkin, J. Phillips, T. Sosnowski, T. Norris, J. Singh, and P. Bhattacharya, "Strain Tensor, Electronic Spectra, and Carrier Dynamcis in In(Ga)As/GaAs Self-Assembled Quantum Dots," Proceedings of 24th International Symposium on Compound Semiconductors, 1997.
- T.S. Sosnowski, T.B. Norris, H. Jiang, J. Singh, K. Kamath, and P.Bhattacharya, "Ultrafast Carrier Capture and Relaxation in InGaAsSelf-Organized Quantum Dots," postdeadline paper, OSA/ILS-XIII, 13th Interdisciplinary Laser Science Conference, Long Beach CA.

#### IV. Scientific Personnel Supported

- 1. Prof. Theodore B. Norris, Principal Investigator
- 2. Timothy Meade, Graduate Student Research Assistant
- 3. Junji Urayama, Graduate Student Research Assistant
- 4. Yun-Shik Lee, Postdoctoral Research Assistant

#### V. Inventions

No patents applied for.

VI. Appendices

Appendix I: Awards

Fellow, Optical Society of America, 2000.

Paul Rappaport Award for best paper in an IEEE Electron Devices Society publication, 1999.

Appendix II: Publications.

Attached please find reprints and preprints of the publications describing the work supported by this program.